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A search for noble-gas evidence for presolar oxide grains

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We present early results from an ongoing search for isotopically distinctive noble gases as evidence for presolar oxide grains. With some qualifications, we do not see such evidence in spinel rich acid residue fractions from the Allende meteorite. We remain hopeful that less abundant mineral separates may yet be fruitful.

Presolar grains, micro-diamonds, silicon carbide, and graphite, have been found in primitive meteorites (1, 2, 3). While the abundances of these three refractory C rich grains are low, a few hundred ppm, a few ppm, &<1ppm respectively in primitive meteorites (4), they are tagged with high concentrations of isotopically anomalous noble gas components, Xe-HL, Kr & Xe-s and Ne-E(H), and Ne-E(L). These tags have served as tracers and allowed the development of techniques for their purification and eventual identification. One might expect similar amounts of refractory presolar oxides to have survived, but so far only three cases exist for their identification. The first two cases are individual corundum oxide grains. Huss *et al.* (5) found one such grain from an Orgueil residue with an $^{26}Al/^{27}Al$ ratio of 8.9 x 10-5, about 18 times higher than the canonical initial solar system value. The second corundum grain, from Murchison, has been found by Nittler *et al.* (6) to have unusual oxygen in addition to a similar $^{26}Al/^{27}Al$ ratio of 8.7 x 10-4 The oxygen was depleted in ^{18}O by 22% and enriched in ^{17}O by a factor of 2. The third case is a measurement by Zinner *et al.* (7) on an aggregate of fine grained spinels from a Murray residue with an ^{17}O enrichment and a possible ^{18}O depletion similar to the second grain, but much subdued. This is consistent with a few such presolar grains diluted by a much larger population of isotopically normal corundum grains and an even larger number of normal spinel grains.

As the three known presolar grains can account for all of the (abundant) anomalous noble gas components found in primitive meteorites, we cannot expect to have the surviving presolar oxides to be so conveniently tagged. What sort of distinguishing compositions can we hope for? Certainly, they are likely to have anomalously high spallation gas concentrations. In addition they may have excess ^{244}Pu fission xenon, or even fission from a shorter lived transuranic element such as ^{248}Cm, although only in grains large enough to trap fission fragments, or in grains occurring in large enough clusters. Such components have already been suggested as possibly detected by Srinivasan *et al.* (8) and by Tang and Anders (9). If one were to concentrate such oxides sufficiently, their trapped noble gas compositions may be distinctive and may serendiptitiosly be abundant enough to stand out. Following the model of the two most abundant known presolar grains, diamond and silicon carbide, refractory and chemically resistant phases are most likely to have survived the formation of the solar system and to be distinctive enough to enable their separation and identification from the bulk of the meteorite. Corundum, as has been found, fits this model. Refractory inclusions have refractory grains in abundance, but appear to have been produced in the early solar system by some high temperature processes, producing partial to nearly complete isotopic homogenization with the average solar system material, as judged by the magnitude of the residual isotopic anomalies found in them (10). However the few presolar silicon carbide grains found in situ in these meteorites were isolated individuals in the matrix, and not found in large aggregates (11). The relatively abundant minerals from the refractory inclusions should therefore just be an (unfortunate) diluent for the presolar grains. The high noble gas concentrations in SiC, which are released in pyrolysis near 1800°C, hinder the detection of noble gases from refractory oxide grains. We have therefore returned to a residue from the Allende C3V meteorite, which has at most very little silicon carbide. Indeed, one goal will be to see if the presence of any silicon carbide can be confirmed.

We have measured He, Ne, Ar, Kr, and Xe, extracted by stepped pyrolysis, on samples prepared from Allende residue BA, a bulk HF-HCl residue. It was treated with perchloric acid twice at 190°C to remove carbon and chromite. Then the presolar diamonds, with their abundant Ne & Xe-HL were depleted >95% six times over (total depletion about 6,000,000) by removal as a colloidal suspension at pH~11. The resulting sample was then divided into 4 size fractions, yielding Allende FF, FG, FH, & FI, nominally >10μm, 3-10μm, 1-3μm & <1μm. Partial data for the last three fractions are given in table 1 and figures 1 and 2. We intend to dissolve the spinel in aliquots of these samples and measure the even more concentrated corundum, hibonite, and possible SiC.

At this stage, and with a couple of caveats, we see no evidence for any presolar grains. First, there is negligible evidence in figure 1 for an isotopic enrichment in ^{22}Ne, as would be carried in SiC. Estimating a conservative upper limit for a ^{22}Ne excess of 10% of the total, table 1, yields an upper limit for SiC of 0.1 ppm, ~60 times less than in Murchison. The spallation Ne, table 1, is more abundant in these fractions than in other Allende samples, but this is expected for nearly pure spinel with its high Mg and Al abundance. Dissolution of this spinel should yield a more sensitive test both for small amounts of SiC and for possible remaining presolar oxide grains carrying excess spallation Ne.

The Xe isotopic composition is consistent with a mixture of trapped Xe, Xe-HL, and ^{244}Pu fission Xe, with no definite evidence for ^{248}Cm fission Xe, though the presence of the He-HL (table 1) in these three samples leaves

some room for argument. The fission rich extraction steps are near 2000°C and should not have Xe-HL remaining. But then these samples should not have any diamonds at all remaining. Yet there is Xe-HL! However, the measured Ne_T/Xe-HL ratio is lower than that of diamonds (table 1.) Hence the Xe-HL cannot be from average presolar diamonds. A distinct carrier leaves open the question of how much contribution Xe-HL makes to the fission rich fractions in figure 2. If those fractions have significant Xe-HL, then the pure fission composition on the abscissa lies to the left of ^{244}Pu, corresponding to a possible contribution from ^{248}Cm.

The concentration of fission Xe in these three fractions lie in the range measured in refractory inclusions (Table 1), consistent with these inclusions being the source of most of these spinels. Any proposed presolar oxides then make negligible contribution to the observed fission Xe. And yet, the high release temperature of the fission Xe, near 2000°C, 500°C above that of the spallation Ne release peak, could be interpreted as an indication that the fission Xe is carried in a minor fraction of the grains, more refractory than the spinel, and at a corresponding higher concentration. If the spallation Ne in each extraction step is used as a measure of the mass sampled (an overestimate of the mass if there are presolar grains with high exposure ages), then the residual spallation Ne in the fission rich steps implies a fission Xe concentration 10 to 20 times higher than found in refractory inclusions. Of course spinel itself melts at >2000°C. If the spinel does not react with the Ta foil container in our extraction furnace, then we may just be seeing diffusive fractionation between the Ne and the Xe.

This work neither finds strong evidence for presolar oxides, nor yet rules out the possibility of their separation.

Table 1		$^{22}Ne_{Spall}$	^{22}NeE(H)	$^{22}Ne_T$ / $^{132}Xe_{Ex}$	$^{132}Xe_{Ex}$	$^{132}Xe_{fiss}$
Sample:		E-8 cc/g	E-8cc/g		E-10 cc/g	E-10 cc/g
Allende:						
bulk	(12)	1.7				
refract. incl.	(13,14)	1.3-1.8				0.10-0.18
diamonds	(15)			400	2500	
FF >10mm		3.20	<0.3	66	0.57	0.13
FG 3-10mm		2.61	<0.3	65	1.57	0.18
FH 1-3 mm		2.76	<0.3	59	3.44	0.17
Murchison:						
SiC average	(16)	23	16,700			

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